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6 HYDROGEN INDUCED INTERGRANULAR CRACKING OF NICKEL-BASE ALLOYS.

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by

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This report is intended to summarize the activities pursued during the first year of a program directed toward an evaluation of the susceptibility of nickel and nickel base alloys to embrittlement by cathodically produced hydrogen. Preliminary results suggest that hydrogen permeation in unstrained membranes occurs by lattice diffusion rather than by alternate short circuiting mechanisms. Work is in progress to evaluate the contribution of dislocation transport to the permeation flux.

1. Introduction

Earlier work [1] has shown that commerically pure nickel tensile specimens are susceptible to intergranular cracking when exposed to cathodically produced hydrogen in aqueous electrolytes at ambient conditions. Cracking was associated with the accumulation of impurities such as Sn, Sb, P and S and the depletion of Cu in the grain boundaries. The segregated elements are known to be poisons for the combination of hydrogen adatoms produced by proton reduction and, hence, stimulate the absorption of hydrogen by the metal. Significantly, this susceptibility may be eliminated if the above grain boundary partitioning is avoided during heat treatment [2]. The intergranular cracking of nickel-base alloys used, for example, as tubing material in pressurized water reactor (PWR) steam generators [3] as well as in oil and gas production [4][5] and chemical processing applications is in many respects phenomenologically similar to that of pure nickel [6]. Hence, the principal aim of this program is to examine the cracking of alloys such as Inconel 600, Incoloy 800, and Hastelloy C-276 polarized cathodically in aqueous electrolytes at ambient conditions in the context that cracking may be induced by hydrogen.

This report is intended to serve as a summary of our activities during the first year of this program. Research in progress is proceeding in essentially two directions: firstly, straining electrode experiments are underway on nickel-base alloys polarized

cathodically in acid and alkaline electrolytes under ambient conditions. At the same time, parallel hydrogen permeation experiments are being performed on membranes of the same alloys having identical metallurgical histories. This report will emphasize the permeation measurements which are furthest along.

Experimental

The basis for the permeation experiments which are being pursued may be understood with the aid of Figure 1. Hydrogen permeation may be presumed to occur by means of lattice diffusion, grain boundary diffusion or, in the case of specimens undergoing plastic deformation, dislocation transport [8][9]. We expect that some hydrogen may be preferentially absorbed into the matrix at grain boundary intersections with the free surface and then travel along such boundaries throughout the bulk. Of course, hydrogen enters the solid at locations other than poisoned grain boundaries, evidence for which is the fact that serrated yielding occurs during tensile deformation of nickel single crystals charged with hydrogen while being deformed [10]. Dislocation-hydrogen interactions have been observed in precharged nickel single crystals as well [11]. Hence, some hydrogen is likely to be dragged into the interior along with mobile dislocations and may accumulate at grain boundaries as suggested in Figure 1. Lattice diffusion in nickel single cyrstals is relatively slow:

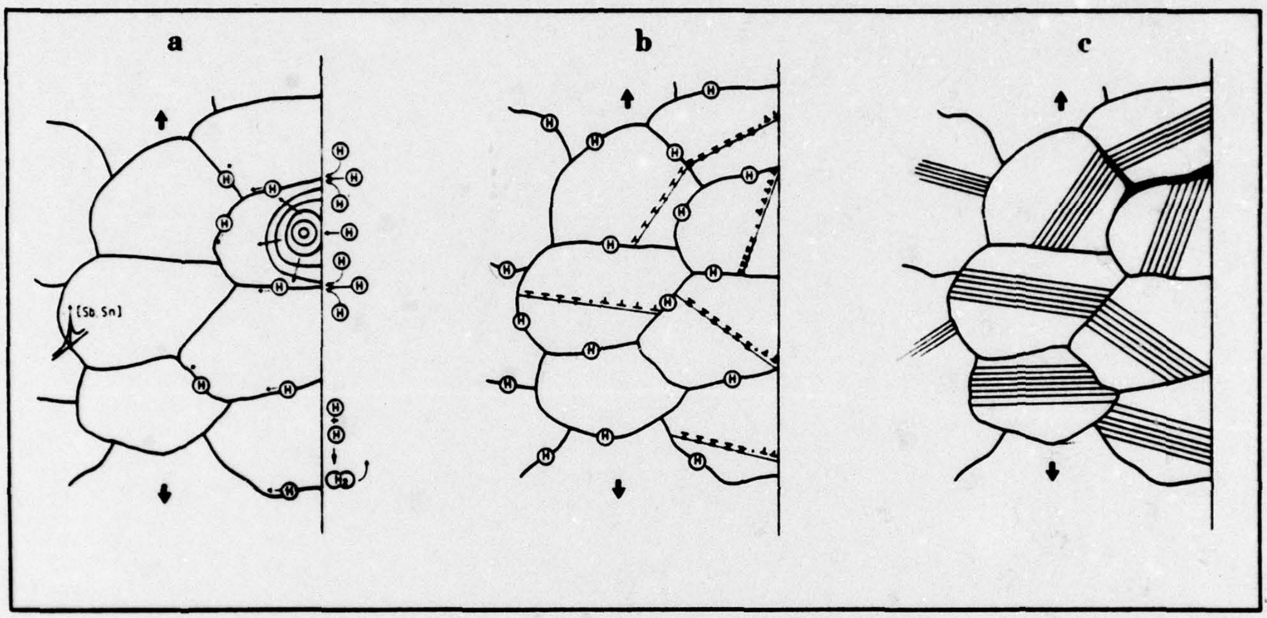

FIGURE 1. Processes likely to be involved in embrittlement [7].

the diffusion coefficient for hydrogen at 25°C is of the order of $10^{-10}cm^2/sec$ [12][13]. The present work is intended to determine the contribution of each such transport process in the permeation of hydrogen through nickel and nickel-base alloys.

The experimental cell which is being used in this work is shown in Figure 2. This cell is of the Devanthan design [14] with the exception that provision has been made to allow the membranes under study to be plastically deformed while being cathodically charged with hydrogen. In this technique, cathodically generated atomic hydrogen is absorbed into a metal specimen (typically 100 - 150 μm thick), travels the thickness and is subsequently oxidized on the anodically polarized exit side, as shown schematically in Figure 3. The hydrogen oxidation current is a direct measure of the rate at which hydrogen permeates the membrane. A typical transient for a palladium membrane is shown in Figure 4. Palladium is perhaps the most hydrogen permeable metal, and serves in this case to demonstrate the operational performance of our permeation cell. Detailed analysis of such transients presents some uncertainties as described by Namboodhiri and Nanis [15] or more recently by Early [16]. Nevertheless, one may calculate with reasonable confidence both the hydrogen diffusion coefficient and the concentration of hydrogen just beneath the cathode surface. In the case of Figure 4, the diffusivity of hydrogen in Pd was determined to be $1.0 \pm 0.3 \times 10^{-7} cm^2/sec$ which compares very well indeed with values reported in the literature for both electrochemical and non-electrochemical measurements.

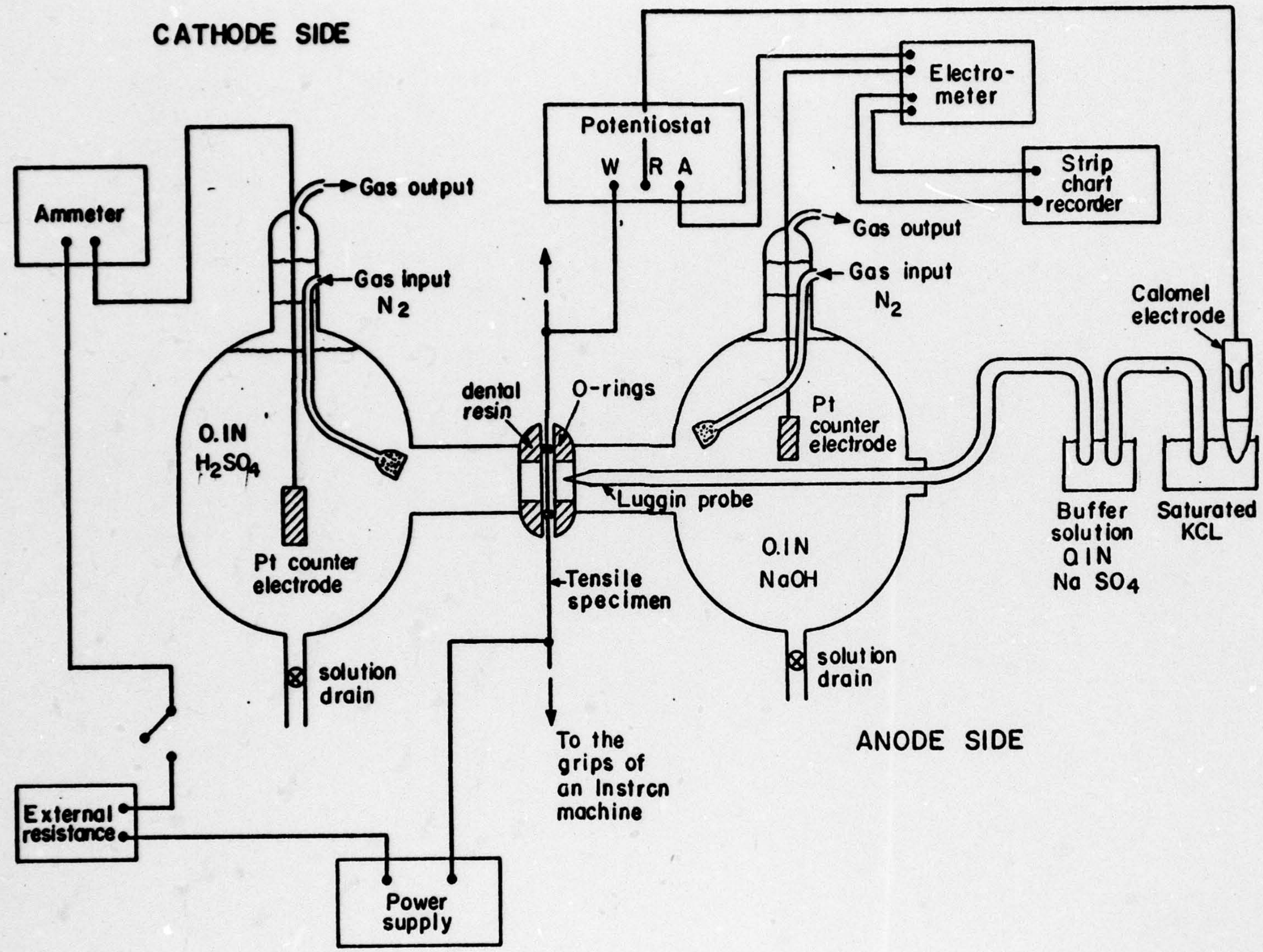

FIGURE 2. Schematic of Devanathan type permeation cell, modified to allow plastic deformation of the specimen while being cathodically charged.

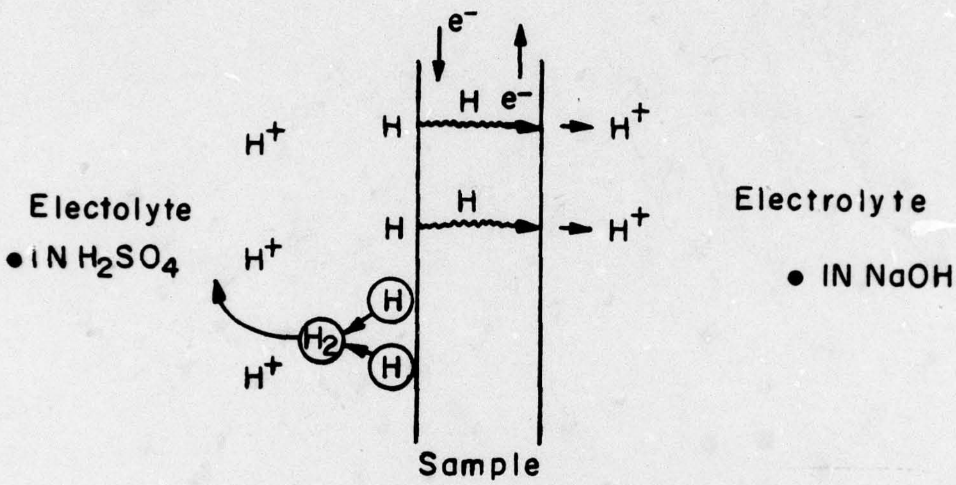

FIGURE 3. The permeation experiment in illustrated format.

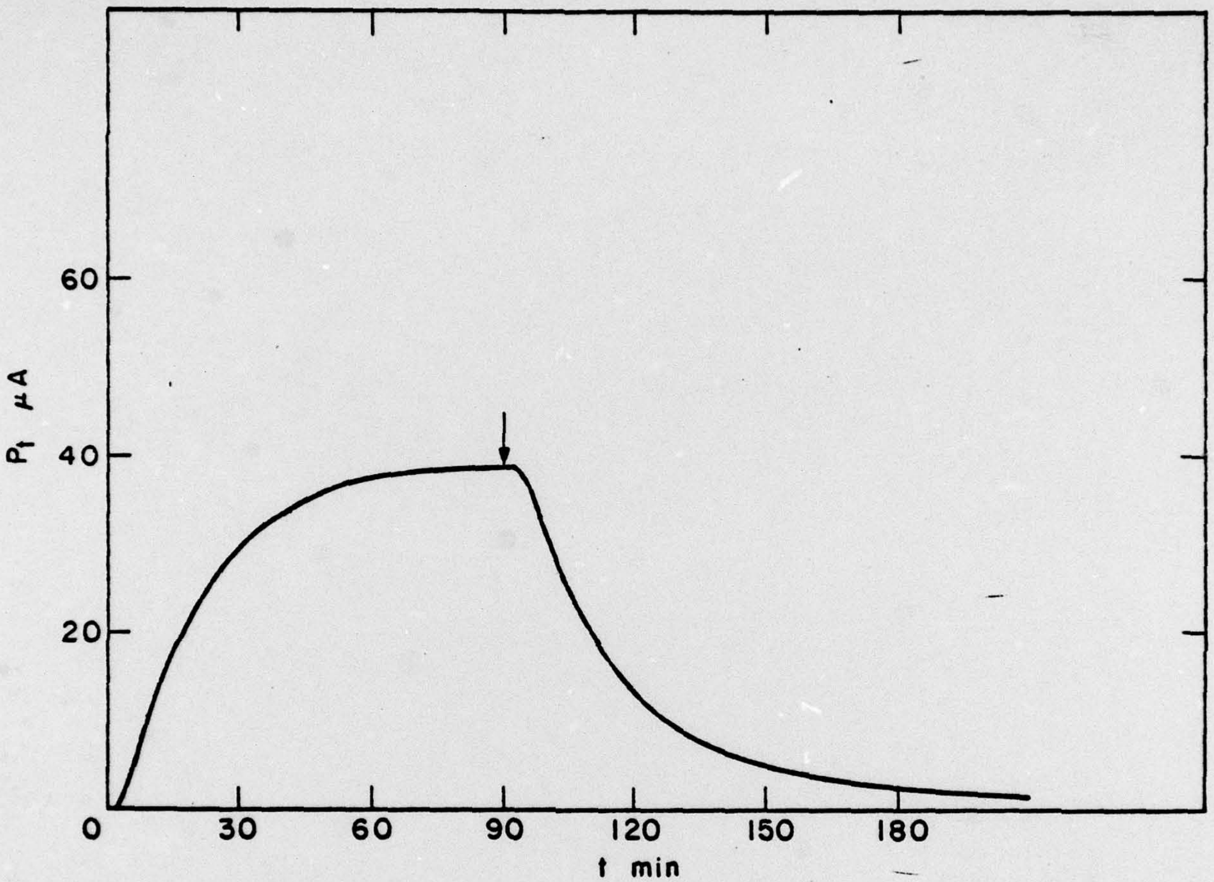

FIGURE 4. Typical permeation transient for an unstrained polycrystalline palladium membrane.

The results of some of the experiments which are in progress on Ni 270 specimens are described in the following section. Additional work which is in progress but not sufficiently advanced at this point to warrant discussion will be treated in subsequent reports.

Results

In the following, we summarize the results of preliminary experiments performed on unstrained membranes with a view to evaluating the grain size dependence of the hydrogen permeation flux. In particular, we have chosen in this case to examine nickel membranes prepared from specimens heat treated similarly to samples found susceptible to embrittlement in earlier work [1][2]. Hence, specimens were encapsulated in the presence of a nitrogen atmosphere and heat treated at 1000°C for various periods of time, thereby producing membranes with grain size in the range 200 - 800μm. Typical rise transients are shown in Figure 5 and 6 and a tabulation of measured diffusion coefficients (D_H), steady state permeation flux (J_∞), and concentration (C_o) is given in Table I.

TABLE I: Summary of Preliminary Permeation Data

Grain Diameter (μm)	J_∞(μA)	C_o(g·atom/cm^3)	D_H(cm^2/sec)
200	0.29	8.55×10^{-5}	6.85×10^{-10}
600	0.15	1.21×10^{-4}	2.40×10^{-10}
800	0.37	3.30×10^{-4}	2.06×10^{-10}

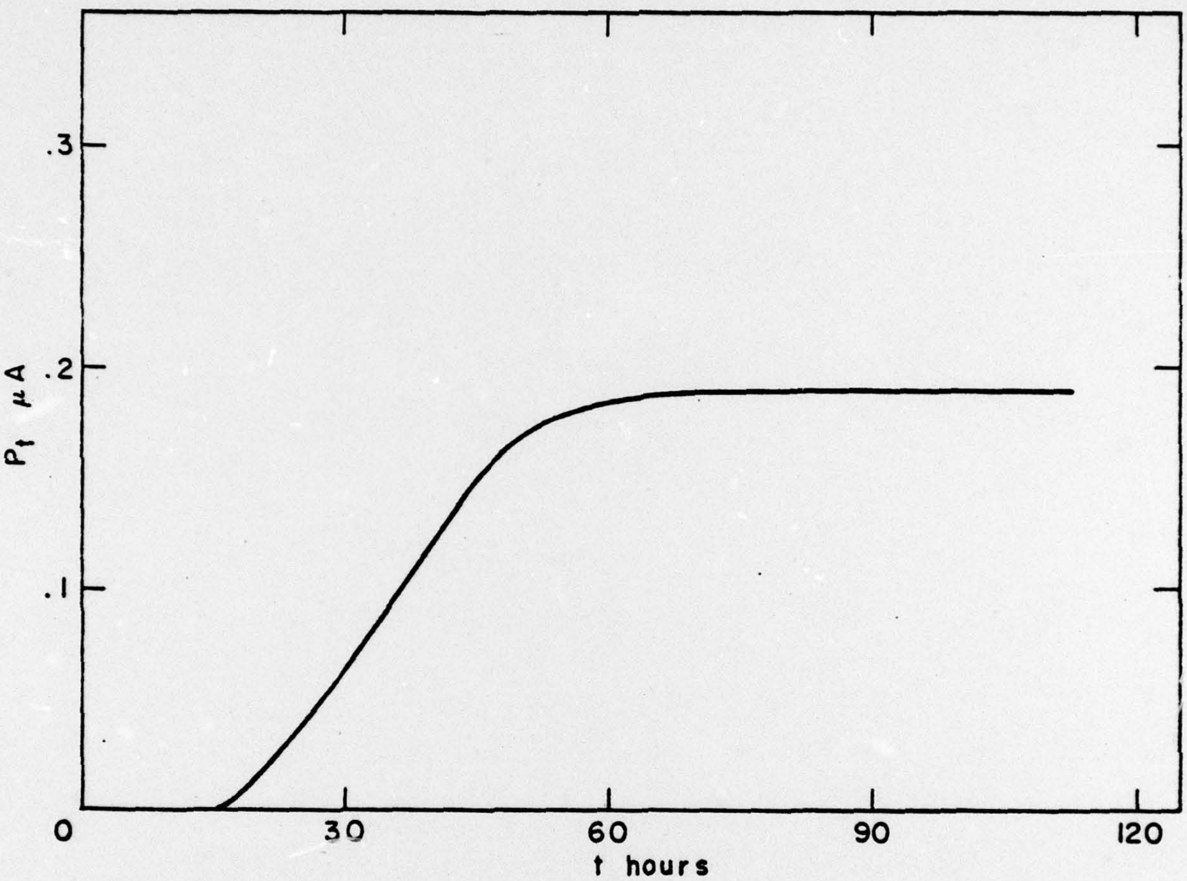

FIGURE 5. Permeation transient for 200μm grain-size nickel membrane. The anodically polarized side of the membrane was electrolessly plated with a thin palladium layer in order to reduce the background oxidation current. Charging current 12.5 mA/cm^2.

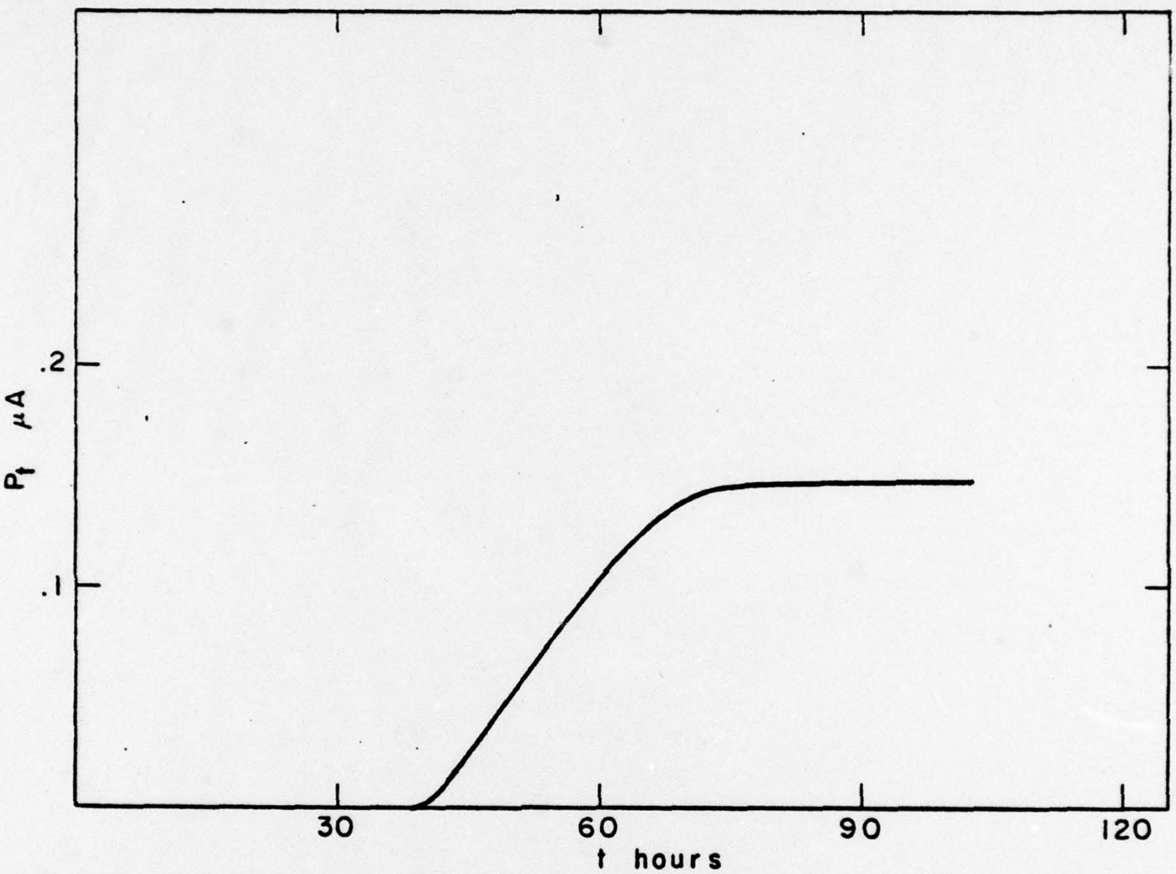

FIGURE 6. Rise transient for 600μm grain-size nickel specimens. Charging current 12.5 mA/cm^2.

It seems to us premature at this stage to comment in detail on these data. It might be worth noting, however, that the diffusion coefficients which have been observed are of a magnitude typical of lattice diffusion and not grain boundary diffusion, the latter of which one might expect to be somewhat higher. Robertson [17] and more recently Fiore et al. [18] have reported somewhat similar observations in permeation experiments performed using molecular and electrochemical hydrogen sources, respectively. This finding, which is still being pursued in our laboratory, suggests by implication that the transport of hydrogen by mobile dislocations may have contributed significantly to the ingress of hydrogen during experiments such as those described earlier [1][2]. We will, of course, examine this hypothesis in detailed experiments utilizing the straining cell which is described in Figure 2. The work will be treated in subsequent reports.

Concluding Remarks

This report is intended to summarize activities pursued during the first year of a program directed toward an evaluation of the susceptibility of nickel and nickel-base alloys to embrittlement by cathodically produced hydrogen. Preliminary results suggest that hydrogen permeation in unstrained membranes occurs by lattice diffusion rather than by alternate short circuiting mechanisms. Work is in progress to evaluate the contribution of dislocation transport to the permeation flux.

References

1. R.M. Latanision and H. Opperhauser, Jr., Met. Trans. 5, 483 (1974).

2. R.M. Latanision and H. Opperhauser, Jr., Met. Trans. 6A, 233 (1975).

3. D. van Rooyen, Corrosion, 31, 327 (1975).

4. R.D. Kane, M. Watkins, D.F. Jacobs, and G.L. Hancock, Corrosion, 33, 309, 1977.

5. R.D. Kane and B.J. Berkowitz, Private Communication, October 1978.

6. R.M. Latanision, O.H. Gastine, and C.R. Compeau, First Technical Report to ONR (January 1978). To be published in Proc. Symposium on Environment Sensitive Fracture of Engineering Materials, Z.A. Foroulis, ed., in press.

7. R.M. Latanision and H. Opperhauser, Jr., Proc. 3rd International Conference on Strength of Metals and Alloys, Vol. 1, 472 (1973).

8. J.K. Tien, A.W. Thompson, I.M. Bernstein and R.J. Richards, Met. Trans., 7A, 821 (1976).

9. H.H. Johnson and J.P. Hirth, Met. Trans., 7A, 1543 (1976).

10. R.M. Latanision and R.W. Staehle, Scripta Met., 2, 667 (1968).

11. A.H. Windle and G.C. Smith, Metal Science Journal, 2, 187 (1968).

12. E. Eichenauer, Mem. Sci. Rev. Met., 57, 943 (1960).

13. Y. Ebisuzaki, W.J. Kass and M. O'Keeffe, J. Chem. Phys., 46, 1378 (1967).

14. M.A. Devanathan and Z. Stachurski, Proc. Roy. Soc., A270, 90 (1962).

15. T. Namboodhiri and L. Nanis, Acta Met., 21, 663 (1973).

16. J.G. Early, Acta Met. 26, 1215 (1978).

17. W.M. Robertson, Z. Metallk., 64, 436 (1973).

18. N.F. Fiore, A. Atrens and D. Mezzanotte, Paper presented at Fall Meeting of AIME, St. Louis, October 1978.

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